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ABSTRACT

In this work, we present a biexciton state population in (111)B oriented site-controlled InGaAs quantum dots (QDs) by resonant two photon excitation. We show that the excited state recombines emitting highly pure single photon pairs entangled in polarization. The discussed cases herein are compelling due to the specific energetic structure of pyramidal InGaAs QDs—an antibinding biexciton—a state with a positive binding energy. We demonstrate that resonant two-photon excitation of QDs with antibinding biexcitons can lead to a complex excitation-recombination scenario. We systematically observed that the resonant biexciton state population is competing with an acoustic-phonon assisted population of an exciton state. These findings show that under typical two-photon resonant excitation conditions, deterministic biexciton state initialization can be compromised. This complication should be taken into account by the community members aiming to utilize similar epitaxial QDs with an antibinding biexciton.

Quantum dots (QDs) are sources of non-classical light—single and entangled photons—a resource that can be used to implement quantum information processing. Practically, a QD has to be excited either optically or electrically to create excitonic complexes occupying the atomic-like energetic levels, which then spontaneously recombine emitting single photons. A specific excitonic complex composed of two pairs of electrons and holes with a net zero spin—a biexciton—recombines in cascaded events[1] emitting a pair of polarization-entangled photons.[2] It is a very attractive resource for quantum information processing; for example, it potentially allows for a simplification of quantum circuit designs due to a reduced number of entangling gates or for innovative heralded schemes.[3] Historically, the typical approach to excite a QD to the biexciton state has been of non-resonant nature (optical or electrical excitation of the QD barriers)—a process that is incoherent, noisy, and largely inefficient due to the probabilistic population of different types of excitonic complexes, overall degrading the properties of quantum light emitters. Recent advancements in quantum optics theory and experimental settings marked a paradigm change toward resonant excitation methods to maximize QD benefits, especially valuable if combined with a QD site-control capability.

By exploiting resonant methods, population inversion of exciton and biexciton states can be achieved via Rabi oscillations,[4,5] adiabatic rapid passage,[6,7] and, later proposed and demonstrated, robust schemes utilizing phonon dressed states.[8–10] Irrespective of being a coherent or an incoherent phonon-assisted process, two-photon excitation is becoming a standard initialization process for the biexciton state in experiments and applications requiring a high quality pair of polarization-entangled photons. Near unity initialization fidelity,[11] spectral purity,[12] near perfect single photon emission,[13] and others, sometimes in combination with recent integrated photonics advancements, are all critical outcomes of these approaches.

Experimentally, the two-photon excitation scheme can be realized because of the non-degeneracy of the exciton and biexciton states arising from the interplay of Coulomb interactions and zero-dimensional geometrical constrictions.[14] The energy difference $\Delta E_B = E_X - E_{XX}$ between the exciton E_X and biexciton E_{XX} is referred to as the binding energy of the biexciton complex. In III–V epitaxial QDs, the ΔE_B value is typically within the range of a few meV. While coherent two-photon excitation (TPE) fulfills the energy conservation requirement $2E_{laser} = E_{XX} + E_X$, the laser photon energy E_{laser} will be

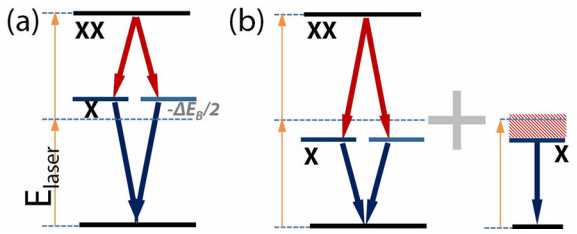

FIG. 1. Two-photon excitation schema in QDs with different types of biexcitons (XX). (a) A binding biexciton two-photon excitation (TPE). (b) An antibinding biexciton TPE competing with the phonon-assisted exciton (X) population.

detuned from the exciton by $-\Delta E_B/2$ [Fig. 1(a)]. Thus, picosecond pulses satisfying the TPE condition can relatively easily address the biexciton state without, for example, the need for cross-polarization filtering to suppress the laser light, which would compromise the polarization state, source intensity, purity, and deterministic initialization.

Noteworthily, the majority of recent reports found in the literature deal with the so-called binding biexciton case ($\Delta E_B > 0$). In a basic picture, higher order excitonic complexes in bulk, two- and one-dimensional structures are always binding, as the binding energy is the result of Coulomb interactions and stable complexes can appear only at lower energy.[15] On the other hand, in zero-dimensional structures, geometrical constraints limit the space available for the charge carriers to redistribute due to Coulomb interaction.[14,16] In specific cases, this can give rise to higher order complexes, so-called antibinding biexcitons, with $\Delta E_B < 0$. Such states have been observed in a number of cases;[15,17–21] however, no experimental study concentrating on the specifications of the antibinding state resonant initialization has been reported so far.

To address an antibinding biexciton by a resonant two-photon excitation, the laser energy has to be detuned positively from an exciton by $\Delta E_B/2$ [Fig. 1(b)]. As we will show later, this can lead to very different QD population dynamics, which, unfortunately, might not necessarily produce a favorable outcome for QDs with an antibinding biexciton in the contest of quantum information processing. Schematically, this population and recombination complexity is outlined in Fig. 1(b)—in parallel to the expected two-photon excitation of a biexciton, favored conditions to excite an exciton state through the interaction with acoustic phonons can occur, i.e., a single photon can give some of its energy to acoustic phonons and utilize the rest to excite only the exciton state.

In our manuscript, we report systematic experimental evidence of this complex dynamics. As a case study, we have selected an $In_{0.25}Ga_{0.75}As/GaAs$ site-controlled pyramidal QD system. All *single* QDs from this QD family exceptionally possess antibinding biexcitons,[22] making them an interesting playground for fundamental research of resonant excitation phenomena. Noteworthily, when two or more identical single $In_{0.25}Ga_{0.75}As$ QDs are stacked with a separator of ≤ 2 nm, they form interacting or coupled-QD complexes with very different energetic structures.[23] Some of them are single QD-like, with a binding or even degenerate biexciton;[24] however, due to small binding energy (0.53 ± 0.47 meV) and higher fine structure splitting values, these QDs are less attractive candidates for resonant TPE experiments.

The QDs have been prepared by metalorganic vapor phase epitaxy (MOVPE) on (111)B-oriented GaAs substrates pre-patterned with 7.5 μm tetrahedrons ensuring site control with the precision of a few nanometers—arguably an important advantage over self-assembled QD systems. $In_xGa_{1-x}As$ QDs with a nominal indium content of 0.25 were confined by GaAs (see the supplementary material for more details). The most notable and attractive feature of these QDs is their high symmetry. An intrinsically highly symmetric (C_{3V} in group theory terms) carrier confinement potential, theoretically foreseen for all (111) direction grown QDs[25,26] ensures a record density of QDs emitting polarization-entangled photon pairs under non-resonant optical[27] and electrical[28] excitation.

QDs have been characterized in a micro-photoluminescence setup at 8 K in a closed-cycle helium cryocooler. Fourier transform-limited 10 ps pulses have been pulse-shaped in a 4f (f = 0.5 m) configuration with a portable tunable-width slit. The base 80 MHz 120 fs pulse width emission was from a tunable Ti:Sapphire laser. The laser emission was filtered with a series of 3 volume Bragg gratings utilized as narrow-band (0.3 nm) notch filters. Spectra have been taken by a CCD paired with a 1 m length monochromator with a 950 mm^{-1} groove density ruled diffraction grating. The second-order correlation measurements[18] were taken by filtering exciton and biexciton transitions with two identical monochromators and utilizing single-photon avalanche photo detectors with dark counts of around 40 per second.

Figure 2(a) shows a QD spectrum ($\Delta E_B = -2.5$ meV) obtained under two-photon excitation tuned resonantly to the biexciton state (several other representative QD spectra are shown in the supplementary material). The observation of the biexciton peak confirmed the possibility to excite QDs with an antibinding state resonantly. Autocorrelation of both, an exciton (X) and biexciton (XX), transitions showed clear antibunching in the second-order (intensity) correlation curves, confirming high-quality single-photon emission from both lines. The measured values of $g^{(2)}(0)$ in the representative case [insets in Fig. 2(a)] were rather low, i.e., 0.03 ± 0.01 (X) and 0.015 ± 0.015 (XX).

Interestingly, we systematically observed clear asymmetries in the intensity of X and XX transitions, indicating substantially different population probabilities of the two states—a rather unexpected result for a state recombining in a cascade event. To understand the origins,

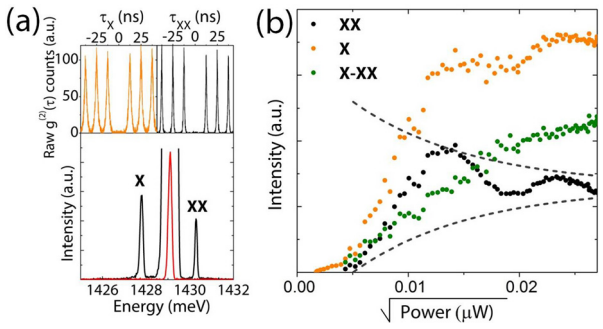

FIG. 2. (a) The spectrum under TPE excitation with 10 ps pulses (the red curve). The insets show the second order auto-correlation curves of the exciton (X) and biexciton (XX) transitions. (b) Excitation power dependence. The dashed lines are the envelopes of the damped Rabi oscillations. The X-XX curve is the difference between the exciton and biexciton intensity at the same excitation power. These are photoluminescence events occurring due to the phonon-assisted exciton population, which do not originate in the biexciton-exciton recombination cascade.

we took the excitation power-dependent measurements presented in Fig. 2(b). Characteristic Rabi oscillations of the excited level (XX) population were observed proving the coherent nature of the process. We attribute the damping to typical dephasing mechanisms, such as dephasing due to phonons at high excitation power.[8] As damping processes typically stabilize the population around 0.5, we estimate that at a π area pulse, the maximum achieved population probability of the biexciton state is 0.65, where the remaining 0.35 population is because of the phonon-assisted exciton initialization discussed below. Small laser energy detuning of $+0.175$ meV and -0.185 meV was sufficient to eliminate the coherent population and shift it toward a phonon-assisted excitation of the biexciton state process (see the supplementary material, Fig. S3).

The exciton, on the other hand, showed a different dependence. While a small oscillation correlated with the biexciton one was observed, an overall emission intensity growth was obtained in the whole power range. This can be predicted in view of the complex mechanism expected for populating an antibinding biexciton case. Specifically, two phenomena are contributing to the intensity of the exciton—(1) photons coming from the biexciton recombination cascade and (2) photons coming from the exciton state that is excited non-resonantly and incoherently[9] through the acoustic phonon bath. The measured intensity is the integrated signal of both. Clear antibunching in the second-order correlation curve suggests that both are taking place during different excitation cycles. To discriminate the contribution of each, we assume, for simplicity, that the exciton population from the cascade is equal to the population of the biexciton. The subtracted result would represent the phonon-assisted X population showing monotonic growth. This is in good agreement with theoretically expected and experimentally observed results for an exciton-only excitation.[8,9] To achieve high population values, this incoherent population process through a phonon-dressed state requires achieving a trade-off between the laser electric field strength and the state preparation time defined by the laser pulse length.[29] This scheme can provide a very robust way of state preparation once realized. The general principle applies for both, the exciton and biexciton, states (in the supplementary material, Fig. 4S shows the phonon-assisted population as a function of laser detuning).

The second-order auto-correlation measurements evidently reveal that the two QD excitation scenarios are competing and do not occur within the same excitation cycle ($g_X^{(2)}(0) = 0.03 \pm 0.01$). It is an expected result as, with a probability dependent on the excitation conditions, a QD is excited either to the exciton or biexciton state—the population of one state blocks the population of the other. Provided that both processes can occur within the timescale of the laser pulse length in general, reexcitation of a QD becomes unlikely and results in low $g^{(2)}(0)$ values of the exciton state. While such a two-channel excitation-recombination mechanism creates deterministic state preparation issues, it does not prevent the detection of photons originated solely during the biexciton recombination cascade. These photons are of primary interest due to the expected polarization entanglement.

To measure polarization entanglement of these pairs of photons, the biexciton photon was used to herald the exciton photon originated in the recombination cascade. By correlating them, second-order cross correlation curves were obtained. Twelve polarization-resolved measurements have been made to obtain full sets of two-photon polarization states in linear, diagonal, and circular bases (Fig. 3). Every

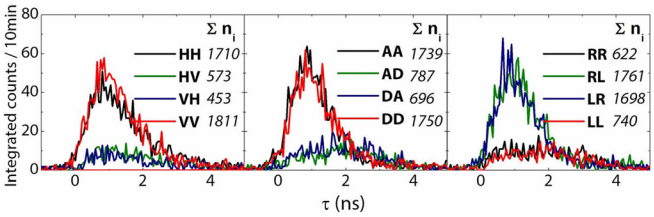

FIG. 3. Polarization entanglement between the photons emitted during the biexciton-exciton recombination cascade. Full measurements in linear, diagonal, and circular polarization bases. The fine-structure splitting of the representative QD is 0.4 ± 0.4 μeV.

individual curve has been integrated for the same duration, i.e., 10 min, to ensure a valid data processing procedure as the raw detection events triggered only by the same excitation pulse have been considered (± 6 ns range). Raw detection counts (given in insets in Fig. 3) were used as the two-photon polarization state intensity (see the supplementary material for the full procedure description).

The biexciton-exciton photon pair entanglement has been estimated by calculating the fidelity to the expected, maximally entangled Bell's state $\frac{1}{\sqrt{2}}(|HH\rangle + |VV\rangle)$,

$$f = \frac{1}{4}(1 + S_{33} + S_{11} - S_{22} + S_{30} + S_{03}), \quad (1)$$

where S_{ii} are the two-photon Stokes parameters.[30] If the QD source is ideal, non-polarized, $S_{30} = S_{03} = 0$, and S_{33}, S_{11}, S_{22} conventionally represent degrees of correlation in linear, diagonal, and circular bases, respectively.[31] In our representative case, we obtained $S_{33} = 0.549$, $S_{11} = 0.403$, $S_{22} = -0.435$, $S_{30} = 0.004$, and $S_{03} = -0.049$, yielding the fidelity value of $f = 0.586 \pm 0.004$. The value exceeding 0.5 is a clear indicator of entanglement ($f \in [0, 1]$), even if the absolute fidelity value is far from recently achieved records.[32] The main entanglement degradation reason can be attributed to the usual cross-dephasing events that destroy the recombination cascade coherence. Indeed, a QD with a very small fine-structure splitting (0.4 ± 0.4 μeV) and pure single photon emission, ideally, should emit pairs with a higher degree of entanglement. However, we systematically observed that these QDs under resonant excitation, in contrast to other types of QDs, maintain a relatively slow (0.78 ns in this specific case) exciton lifetime (see the supplementary material for the lifetime extraction), which is sufficient to increase a cross-dephasing probability substantially.[33] One of the most prominent decoherence mechanisms can be attributed to processing-induced defect states within the bandgap, which can be excited even with the laser tuned to the TPE resonance. The presence of such charge trapping states and destructive effects on optical properties of QDs from the same sample have been demonstrated previously.[22] In agreement with this, a significant linewidth broadening— by spectral wandering—of the exciton (160 μeV) and biexciton (117 μeV) in the here discussed representative case even under TPE proves the existence of a dynamically charged QD environment. Moreover, and in general, under non-resonant excitation conditions, these trapping states are also the main reason for poor single photon emission, linewidth broadening, and QD charging. Thus, results obtained under non-resonant excitation appeared of little interest in the context of this work, and they were not taken for comparison.

Systematic polarization-resolved measurements revealed that the polarization states of photons emitted due to direct phonon-assisted excitation are strongly dependent on the polarization state of the laser. While being the matter of a broader dedicated study in the future, our preliminary observations suggest that the laser polarization state can be mapped on the polarization state of the exciton photon depending on the QD's energetic structure. A demonstration of the phenomenon using the same representative QD as above is shown in Fig. 4. TPE is typically performed using a linearly polarized laser to overcome Pauli exclusion principle[5] (in the supplementary material, we show that the biexciton excitation channel can be effectively suppressed with circularly copolarized photons). Such excitation can efficiently excite the conventional biexciton state, which then recombines in a cascade. The XX curve in Fig. 4 represents these events. The distribution of linear polarization components here has been analyzed by rotating a half-waveplate placed in front of a fixed polarizer at the entrance of the monochromator. Small modulation in the XX intensity is characteristic of this QD and is in good agreement with the two-photon Stokes parameters' measurements, showing that this QD source is slightly polarized. We tentatively attribute this polarization dependence to a weak mixing of heavy and light holes,[34] which can vary in different QDs. The intensity of X events exceeding XX can be assigned to the phonon-assisted initialization of the exciton state (which will refer to as X-XX, i.e., X minus XX). In the case depicted in Fig. 4, these events are fully linearly polarized. The analysed laser intensity is also plotted to show that the polarization state of X-XX photons is nearly identical to the laser's one. The second-order auto-correlation measurement of the X transition [the antibunching $g^{(2)}(\tau)$ curve shown in the inset of Fig. 4] clearly confirmed that the source is fully described by a sub-Poissonian statistics, and it is not laser-related. By rotating the laser polarization by 45° (not shown), the X-XX maximum also shifted, with a just slightly different phase offset from the one visible in Fig. 4, confirming the existence of a correlation function between the polarization states of the laser and X photons created by phonon-assisted excitation.

We speculate that the existence of the polarization phase offset between the maximum of the laser and X-XX photons is related to the energetic fine structure of a QD. A possible scenario suggests that during the excitation moment, the laser polarization state is mapped on to the Bloch sphere representing the exciton spin state.[35–37] In an ideally symmetric QD without the fine-structure splitting, the laser state should be mapped to the polarization state of the emitted photon. However, in the presence of a fine-structure splitting, the exciton spin state, depending on the initial state, can start to precess. The precession mapped on the exciton photon polarization state can be, in principle, resolved by fast single-photon detectors, which unfortunately we do not have access to. In our experiment, we have been integrating emission events without time resolution. A slow precession of the exciton state could be used to explain the observed offsets between the maximum of the laser and X-XX photon events. This scenario should be tested by tuning the fine-structure splitting of the same QD (for example by strain[38]) and performing the same static and time-resolved polarization sensitive measurements.

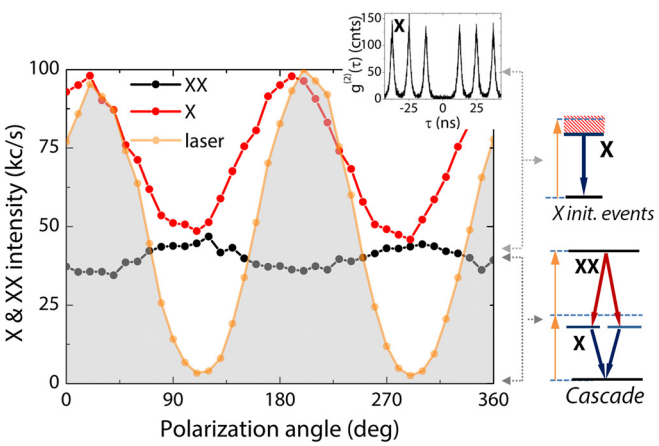

FIG. 4. Linear polarization analysis of the detected light originating from the laser, exciton, and biexciton. The inset shows an antibunching curve collected from the exciton transition.

While it might seem that an antibinding biexciton is a fundamental limitation in the resonant TPE approach, as true deterministic generation of non-classical light becomes compromised, it is worth pointing to a single reported case in the literature,[39] which suggests that under certain conditions, the TPE process actually can be the only excitation phenomenon (no phonon-assisted excitons) despite an antibinding biexciton. It is possible indeed that the TPE process is strongly affected by QD coupling to a cavity or it is highly dependent on the excitation conditions; as in Ref. 39, the authors do not discuss nor observe any issue with the excitonic configuration. A known phenomenon of a strongly reduced excitation power needed to populate cavity-coupled QDs has been observed in the same type of device (200 times reduction in comparison to QDs in a slab).[40] It is possible that a weak laser field strength, which, in general, is driving an exciton coupling to the acoustic phonon bath and is not sufficient to populate the exciton state efficiently during the few picoseconds of the excitation. To achieve a significant phonon-assisted exciton state population with a weak laser field, long preparation times (pulse duration), far exceeding the demonstrated very short lifetime (66 ps) of the biexciton, would be required.[29,41] That kind of microcavity-based device driven by short (a few ps) pulses, in principle, is expected to be robust against phonon-assisted excitation under TPE conditions. Experimental evidence, even though with a binding biexciton, shows that a reduced laser pulse length (from 13 ps to 7 ps) does not affect the state population by resonant TPE; however, short pulses are less efficient in the phonon-induced biexciton state population.[42] We, unfortunately, could not reliably verify this in our experiment due to a rather small binding energy (−2 meV) and the high excitation powers needed, which were causing problems due to stray laser light.

In summary, we have demonstrated the population of site-controlled pyramidal QD biexciton states by two-photon excitation. High-quality single photon emission and good polarization entanglement between the photons emitted through the biexciton recombination cascade were measured. In all the presented cases, we have been dealing with antibinding biexcitons. We showed that this specific fine-structure QD configuration leads to a complex excitation-recombination scenario. It was found that the laser tuned close to a two-photon resonance could populate the exciton state through the acoustic phonon sideband. The latter excitation is less significant at low excitation powers, specifically at the exact resonance condition with a π area pulse. However, as the two phenomena are competing, the true deterministic photon emission is compromised, at least in our $In_{0.25}Ga_{0.75}As$ QD system. On the other hand, there is no obvious

reason why this phenomenon should not be observed in the other types of epitaxial QDs under similar and thus at the moment typical, excitation conditions. The community members aiming to utilize QDs with antibinding biexcitons should be taking into account the findings presented in this work. To exploit the full potential of our highly symmetric site-controlled QDs, a different resonant excitation approach or conditions might be needed. Among possible candidates are phonon-assisted two-photon excitation energetically tuned above the biexciton transition[29] (the feasibility of this experiment is shown in the supplementary material, Fig. 4S) and a variation of dichromatic wavelength excitation.[43,44] Also, a comprehensive study of different excitation conditions, such as the pulse length, pulse area, and effect of a microcavity, could provide the solution to the compromised deterministic biexciton initialization. A here highlighted case presented by Wang *et al.*[39] is in agreement with the theory of phonon-assisted state preparation,[29,41] both suggesting that the phonon-induced population can be suppressed or minimized by designing devices that can be driven by low power and short (a few picoseconds) pulses. Finally, we observed strong correlations between polarization states of the laser and the exciton populated directly with phonon assistance. Our preliminary findings show that the laser state can be mapped on the exciton photon and will be the subject of further studies.

See the supplementary material for sample fabrication and other resonant optical measurement details.

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DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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